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Anion incorporation in AlGaAsSb alloys grown by MBE

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Abstract. An influence of MBE growth parameters (substrate temperature, total group V flux, and As/Sb flux ratio) on a composition of $\text{Al}_x\text{Ga}_{1-x}\text{As}_y\text{Sb}_{1-y}$ alloys ($x = 0.5$) as well as their structural quality have been studied in details. Using these data, As and Sb incorporation coefficients have been calculated in two different ways. An effect of the unintentional Sb incorporation in InAs layers is also discussed.

AlGaAsSb alloys are widely used as cladding layers of laser diodes emitting in mid-infrared spectral range [1]. Such lasers are of great importance for a large variety of applications, including remote trace-gas sensing, pollution monitoring, molecular spectroscopy [2] and optical communications using fluoride glass fibers [3].

One of the general problems in molecular beam epitaxy (MBE) growth, that has not been completely solved so far, is a compositional control of the multicomponent alloys with two volatile elements, in particular, the AlGaAsSb alloys involving As and Sb [4–6]. The available theoretical models of the MBE growth do not allow an unambiguous quantitative description of the composition dependence on growth parameters. An adequate thermodynamic model of anion-mixed alloy MBE growth is still questionable. The difficulties in composition control of the anion-mixed alloys are due to non-unity incorporation coefficients of the volatile elements. Most of the previous papers on MBE of III-(Sb, As) alloys consider situations when both antimony and arsenic cracking cells [4, 5] or the arsenic cracking cell in combination with a conventional antimony cell [6] are used as molecular beam sources. This paper reports on MBE growth of AlGaAsSb alloys using conventional arsenic and cracking antimony cells.

Three series of samples were grown on a RIBER 32P setup equipped with the antimony cracking cell RB-075-Sb providing mostly Sb_2 flux. Both InAs and GaSb substrates were used. The structures contain a 0.2 μm -thick InAs or GaSb buffer layer followed by a (0.5–1) μm -thick $\text{Al}_{0.5}\text{Ga}_{0.5}\text{As}_y\text{Sb}_{1-y}$ layer. The samples of series A were grown at different As fluxes and other equal growth conditions. In the samples of series B and C the varying parameters were the Sb flux and the substrate temperature, respectively, while the As flux was kept constant and high enough to provide $\text{As}/\text{III} > 1$ conditions.

The AlGaAsSb composition was measured by electron probe microanalysis (EPMA) and verified through the simulation of high resolution X-ray diffraction (XRD) rocking curves using the EPMA data on Al and Ga content. Additionally, the compositional profiles of the structures were obtained by SIMS.

Figure 1 presents XRD rocking curves of one of the InAs/AlGaAsSb structures. Three main peaks are clearly resolved in both curves. Besides a dominating InAs substrate peak, there is an intensive peak located at ~ 100 arcsec, which is attributed to the AlGaAsSb layer nearly lattice-matched to InAs ($\Delta a/a < 5 \times 10^{-4}$). The small difference in FWHM values of the corresponding peaks for both ω - and $\theta - 2\theta$ rocking curves as well as an

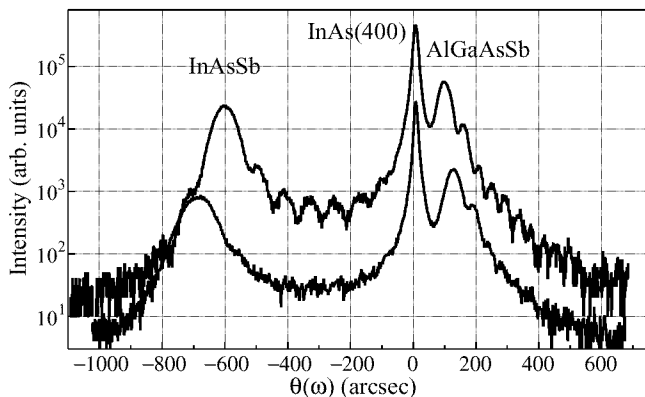


Fig. 1. The (004) ω - (bottom) and $\theta-2\theta$ (upper) XRD rocking curves of the InAs/ $\text{Al}_{0.5}\text{Ga}_{0.5}\text{As}_{0.13}\text{Sb}_{0.87}$ structure.

existence of clearly visible interference fringes confirm the high structural quality and plain morphology of this layer. The third weaker peak at $\sim(620-680)$ arcsec, that is absent in the samples grown on GaSb substrates as well as grown using conventional Sb cell, seems to be attributed to the InAs buffer layer contaminated with Sb. The layer thickness estimated from the period of the third $\theta-2\theta$ peak fringes is consistent well with that of the buffer layer. The presence of Sb in the buffer layers was confirmed by SIMS measurements. An estimation of the $\text{InAs}_{1-y}\text{Sb}_y$ alloy composition from the XRD peak position gives $y \sim 0.05$. The similar effect of the uncontrolled As incorporation into Sb-based compounds was observed earlier [7]. In our case, the main reason of the unintentionally Sb incorporation into As-based layers is probably a significantly increased reactivity of Sb_2 dimer molecules as compared to Sb_4 ones.

It was found from EPMA studying of the samples of series A, that As flux variation within a wide range does not affect the composition of AlGaAsSb, provided that it is grown at the Sb_2 flux large enough to keep V-stabilized growth conditions. The results of EPMA measurements of the samples of series B and C are summarized in Fig. 2 and Fig. 3, respectively. One should note that the samples grown at high growth temperature ($T_S \sim 520^\circ\text{C}$) and high V/III ratio (high As flux) demonstrate much worse morphology and structural quality. The minimum beam equivalent pressure (BEP) of Sb flux corresponding to Sb-stabilized growth of $\text{Al}_{0.5}\text{Ga}_{0.5}\text{Sb}$ was 1.7×10^{-6} and 2.2×10^{-6} Torr at T_S of 480°C and 520°C , respectively, as shown by vertical dashed lines in Fig. 2.

Some of the $\text{Al}_{0.5}\text{Ga}_{0.5}\text{As}_y\text{Sb}_{1-y}$ layers were grown with the composition inside a thermodynamic miscibility gap ($y = 0.3$ and 0.5). The possibility of MBE growth of $\text{Al}_x\text{Ga}_{1-x}\text{As}_y\text{Sb}_{1-y}$ alloys lattice-matched to InP (also inside the thermodynamic instability region) with good structural and optical properties has been recently demonstrated [4]. On the other hand, the authors of Ref. [5] could not penetrate deep in the miscibility gap and assumed that the Al(As,Sb) miscibility gap manifests itself in a shift in As composition due to a rejection of excess As, rather than in phase separation. Therefore, the reliability of our data for $y > 0.3$ does not seem to be high.

In contrast to thermodynamic predictions, the experimental data demonstrate the dominant Sb incorporation in AlGaAsSb layers even at rather low Sb fluxes. Such incorporation behavior of the volatile elements can be explained by a strong difference in kinetically controlled As and Sb incorporation coefficients, defined as a ratio of a number of atoms incorporated into the grown layer to a total number of incident atoms. Thus, to be correct any

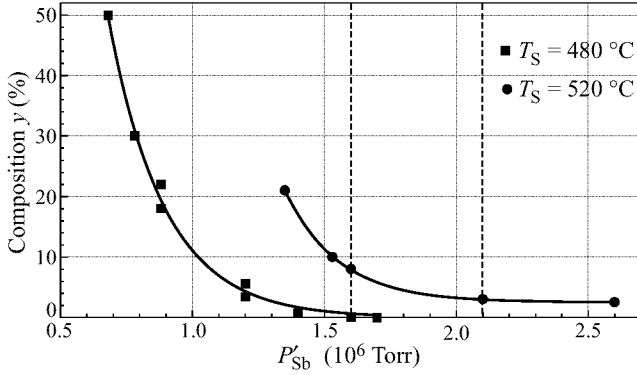


Fig. 2. As content as a function of Sb BEP at two different growth temperatures.

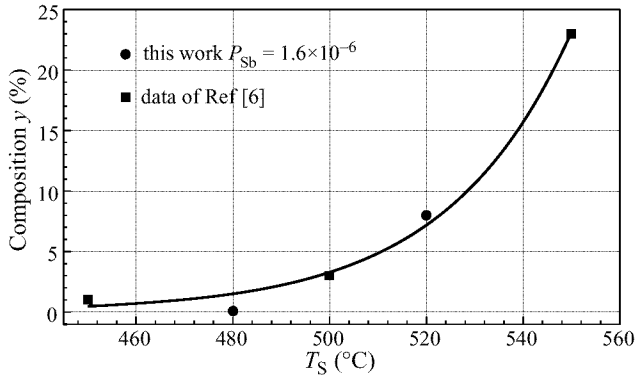


Fig. 3. As content as a function of the growth temperature.

thermodynamic model of MBE should take into account the kinetics processes on growth surfaces.

The analysis of the AlGaAsSb composition dependencies was carried out within the framework of kinetic conception only, using expression for the As and Sb incorporation coefficients as the main parameters affecting the alloy composition.

The dependence of composition on a flux ratio, taking into account the incorporation coefficients of the elements, can be written as follows

$$y = \left[1 + \frac{1}{2\alpha_{As}^N} \frac{\eta_{As_4}}{\eta_{Sb_2}} \left(\frac{P'_{Sb_2}}{P'_{As_4}} \right) \right]^{-1} \quad (1)$$

where $\alpha_{As}^N = \alpha_{As}/\alpha_{Sb}$ is a relative As incorporation coefficient, P'_i is a BEP of the respective molecular fluxes and η_i is a sensitivity coefficient of the Bayard–Alpert ion gauge [8].

To calculate the absolute incorporation coefficients of the elements, the well-known expression for the fluxes condensed on a substrate has been used [9]. Then the As and Sb incorporation coefficients can be expressed as

$$\alpha_{As} = \frac{y v_g \pi r^2 \sqrt{2\pi R M_{As_4} T_{As}}}{a^3 S_{As_4}^{ev} P_{As_4} S_S \cos(\varphi) \sin(\Theta) N_A}, \quad \alpha_{As} = \frac{2(1-y) v_g \pi r^2 \sqrt{2\pi R M_{Sb_2} T_{Sb}}}{a^3 S_{Sb_2}^{ev} P_{Sb_2} S_S \cos(\varphi) \sin(\Theta) N_A} \quad (2)$$

where r is a distance between substrate and cell orifice, φ is an angle between the direction of molecular beam and the normal to cell aperture, Θ is an incident angle; N_A is the

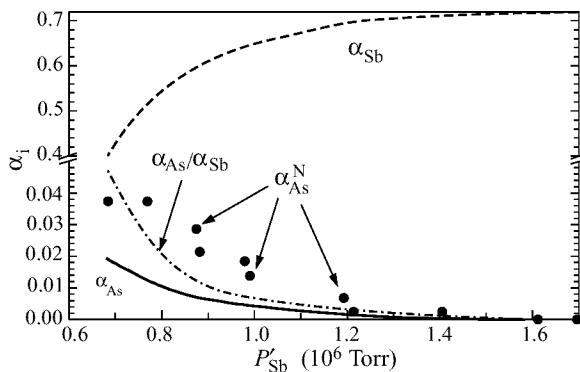


Fig. 4. As and Sb absolute and relative incorporation coefficients versus Sb BEP.

Avogadro number, M_i is molar mass of evaporated element, S_i^{ev} and S_s are areas of a cell orifice and a substrate, P_i is an equilibrium pressure in the cell, v_g is the alloy layer growth rate and a is a lattice constant.

As a result the calculated dependencies of As and Sb incorporation coefficients on Sb BEP are presented in Fig. 4. As shown in Fig. 4, the dependencies of relative As incorporation coefficient on Sb BEP calculated by two different ways, using expression (1) and expressions (2), are in good agreement. A small discrepancy appears to be due to some inaccuracy in estimation of equilibrium pressures and in cell temperature measurements using a thermocouple. The absolute values of As and Sb incorporation coefficients have been found to be much smaller than unity, with the α_{As}/α_{Sb} ratio exceeding 10.

In summary, both the experimental data and theoretical calculations demonstrate strong dependencies of AlGaAsSb alloy composition on growth temperature and Sb flux. The alloy composition has been found to be efficiently controlled by varying only the Sb BEP with Al, Ga, As BEPs and growth temperature kept constant.

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